Higher order antibunching is not a rare phenomenon

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Abstract

Since the introduction of higher order nonclassical effects, higher order squeezing has been reported in a number of different physical systems but higher order antibunching is predicted only in three particular cases. In the present work, we have shown that the higher order antibunching is not a rare phenomenon rather it can be seen in many simple optical processes. To establish our claim, we have shown it in six wave mixing process, four wave mixing process and in second harmonic generation process.

1 Introduction

Antibunching and squeezing do not have any classical analogue [1-3]. Higher order extensions of these nonclassical states have been introduced in recent past [??-7]. Among these higher order nonclassical effects, higher order squeezing has already been studied in detail [??, 5, 8, 9] but the higher order antibunching (HOA) is not yet studied rigourously. The idea of HOA was introduced by Lee in a pioneering paper [6] in 1990, since then it is predicted in two photon coherent state [6], trio coherent state [10] and in the interaction of intense laser beam with an inversion symmetric third order nonlinear medium [11]. From the fact that in last 15 years HOA is reported only in three particular cases, HOA appears to be a very rare phenomenon. The present study aims to establish that this apparent rarity is not due to any physical reason. To establish that, we have shown the existence of HOA in six wave mixing process, four wave mixing process and in second harmonic generation process.

Using the negativity of P function [1], Lee introduced the criterion for HOA as

$$R(l,m) = \frac{\left\langle N_x^{(l+1)} \right\rangle \left\langle N_x^{(m-1)} \right\rangle}{\left\langle N_x^{(l)} \right\rangle \left\langle N_x^{(m)} \right\rangle} - 1 < 0, \tag{1}$$

where N is the usual number operator, $\left\langle N^{(i)} \right\rangle = \langle N(N-1)...(N-i+1)\rangle$ is the ith factorial moment of number operator, $\langle\rangle$ denotes the quantum average, l and m are integers satisfying the conditions $l \leq m \leq 1$ and the subscript x denotes a particular mode. Ba An [10] choose $m = 1$ and reduced the criterion of lth order antibunching to

$$A_{x,l} = \frac{\left\langle N_x^{(l+1)} \right\rangle}{\left\langle N_x^{(l)} \right\rangle \langle N_x \rangle} - 1 < 0 \tag{2}$$

or,

$$\left\langle N_x^{(l+1)} \right\rangle < \left\langle N_x^{(l)} \right\rangle \langle N_x \rangle. \tag{3}$$

Physically, a state which is antibunched in lth order has to be antibunched in $(l-1)th$ order. Therefore, we can further simplify (3) as

$$\left\langle N_x^{(l+1)} \right\rangle < \left\langle N_x^{(l)} \right\rangle \langle N_x \rangle < \left\langle N_x^{(l-1)} \right\rangle \langle N_x \rangle^2 < \left\langle N_x^{(l-2)} \right\rangle \langle N_x \rangle^3 < ... < \langle N_x \rangle^{l+1} \tag{4}$$

and obtain the condition for $l-th$ order antibunching as

$$d(l) = \left\langle N_x^{(l+1)} \right\rangle - \langle N_x \rangle^{l+1} < 0. \tag{5}$$

This simplified criterion (5) coincides exactly with the physical criterion of HOA introduced by Pathak and Garica [11]. Here we can note that $d(l) = 0$ and $d(l) > 0$ corresponds to higher order coherence and higher order bunching (many photon bunching) respectively. As we have already mentioned, higher order antibunching is not yet studied rigourously and apparently, higher order antibunching is very rare since it is reported only in three particular cases in last 15 years. The present work aims to show that its not really a rare phenomenon rather it can be seen in many simple optical processes. To establish that we have used the criterion (5) and short time approximated solutions of equation of motions corresponding

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to various Hamiltonians (such as six wave mixing process, four wave mixing process and second harmonic generation) and have shown that HOA can be seen in all the physical systems selected for the present study. In the next section we present a second order operator solution of the equation of motion of six wave mixing process in detail and use that to show the existence of HOA in six wave mixing process. In section 3 and 4 we have studied the possibilities of observing higher order antibunching in four wave mixing process and in second harmonic generation process respectively. Finally section 5 is dedicated to conclusions.

2 Six wave mixing process

Six wave mixing may happen in different ways. One way is that two photon of frequency ω_1 are absorbed (as pump photon) and three photon of frequency ω_2 and another of frequency ω_3 are emitted. The Hamiltonian representing this particular six wave mixing process is

$$H = a^\dagger a\omega_1 + b^\dagger b\omega_2 + c^\dagger c\omega_3 + g(a^{\dagger 2}b^3c + a^2b^{\dagger 3}c^\dagger) \tag{6}$$

where a and $a^\dagger$are creation and annihilation operators in pump mode which satisfy $[a, a^\dagger]$=1, similarly b, $b^\dagger$ and c, $c^\dagger$ are creation and annihilation operators in stokes mode and signal mode respectively and g is the coupling constant. Substituting $A = a\,e^{i\omega_1 t}, B = b\,e^{i\omega_2 t}$ and $C = c\,e^{i\omega_3 t}$ we can write the Hamiltonian (6) as

$$H = A^\dagger A\omega_1 + B^\dagger B\omega_2 + C^\dagger C\omega_3 + g(A^{\dagger 2}B^3C + A^2B^{\dagger 3}C^\dagger) \tag{7}$$

Since the Hamiltonian is known, we can use Heisenberg's equation of motion (with $\hbar = 1$):

$$\dot{A} = \frac{\partial A}{\partial t} + i[H, A] \tag{8}$$

and short time approximation to find out the time evolution of the essential operators. From equation (7) we have

$$[H, A] \;\; = \;\; -A\omega_1 - 2gA^\dagger B^3C. \tag{9}$$

From (8) and (9) we have

$$\dot{A} = iA\omega_1 - iA\omega_1 - i2gA^\dagger B^3C = -2igA^\dagger B^3C. \tag{10}$$

Similarly

$$\dot{B} = -3igA^2B^{\dagger 2}C^\dagger \tag{11}$$

and

$$\dot{C} = -igA^2B^{\dagger 3}. \tag{12}$$

We can find the second order differential of A using (8) and (10-12) as

$$\begin{aligned} \ddot{A} &= \tfrac{\partial \dot{A}}{\partial t} + i[H, \dot{A}] \\ &= 4g^2AB^{\dagger 3}B^3C^\dagger C - 18g^2A^\dagger A^2B^{\dagger 2}B^2C^\dagger C - 36g^2A^\dagger A^2B^\dagger BC^\dagger C \\ &- 2g^2A^\dagger A^2B^{\dagger 3}B^3 - 18g^2A^\dagger A^2B^{\dagger 2}B^2 - 36g^2A^\dagger A^2B^\dagger B \\ &- 12g^2A^\dagger A^2C^\dagger C - 12g^2A^\dagger A^2. \end{aligned} \tag{13}$$

Now by using the Taylor's series expansion

$$f(t) = f(0) + t\left(\frac{\partial f(t)}{\partial t}\right)_{t=0} + \frac{t^2}{2!}\left(\frac{\partial^2 f(t)}{\partial t^2}\right)_{t=0} \ldots\ldots \tag{14}$$

and substituting (10) and (13) in (14) we get

$$\begin{aligned} A(t) &= A - 2igtA^\dagger B^3C \\ &+ g^2t^2 \left[2AB^{\dagger 3}B^3C^\dagger C - 9A^\dagger A^2B^{\dagger 2}B^2C^\dagger C - 18A^\dagger A^2B^\dagger BC^\dagger C\right. \\ &- \left.A^\dagger A^2B^{\dagger 3}B^3 - 9A^\dagger A^2B^{\dagger 2}B^2 - 18A^\dagger A^2B^\dagger B - 6A^\dagger A^2C^\dagger C - 6A^\dagger A^2\right] \end{aligned} \tag{15}$$

or,

$$\begin{aligned} A(t) &= A - 2igtA^\dagger B^3C \\ &+ g^2t^2 \left[2AB^{\dagger 3}B^3N_c - 9N_AAB^{\dagger 2}B^2N_C - 18N_AAN_BN_C\right. \\ &- \left.N_AAB^{\dagger 3}B^3 - 9N_AAB^{\dagger 2}B^2 - 18N_AAN_B - 6N_AAN_C - 6N_AA\right] \end{aligned} \tag{16}$$

where $N_A = A^\dagger A,\;\; N_B = B^\dagger B,\;\; N_C = C^\dagger C$[4]. The Taylor series is valid when t is small, so this solution is valid for a short time and that is why it is called short time approximation. The above calculation is shown in detail as an example. Following

[4]A short time approximated expression of time evolution of annihilation operator in pump mode of six wave mixing process described by (6) is also derived in [9] but unfortunately their solution contain some mistakes.

the same prescription, we can find out the time evolution of B and C or any other creation and annihilation operator that appears in the Hamiltonian of matter field interaction. This is a very strong technique since this straight forward prescription is valid for any optical process where interaction time is short. After obtaining the analytic expression for time evolution of annihilation operator, now we can use it to check whether it satisfies condition (5) or not.

Let us start with the possibility of observing first order antibunching. From equation (15), we can derive expressions for $N(t)$ and $N^{(2)}(t)$ as

$$\begin{array}{lcl} N(t) & = & A^{\dagger}A-2igt\left(A^{\dagger 2}B^{3}C-A^{2}B^{\dagger 3}C^{\dagger}\right) \\ & + & g^{2}t^{2}\left[8A^{\dagger}AB^{\dagger 3}B^{3}C^{\dagger}C-18A^{\dagger 2}A^{2}B^{\dagger 2}B^{2}C^{\dagger}C-36A^{\dagger 2}A^{2}B^{\dagger}BC^{\dagger}C-2A^{\dagger 2}A^{2}B^{\dagger 3}B^{3}\right. \\ & - & \left.18A^{\dagger 2}A^{2}B^{\dagger 2}B^{2}-36A^{\dagger 2}A^{2}B^{\dagger}B-12A^{\dagger 2}A^{2}C^{\dagger}C+4B^{\dagger 3}B^{3}C^{\dagger}C-12A^{\dagger 2}A^{2}\right] \end{array} \tag{17}$$

and

$$\begin{array}{lcl} N^{(2)}(t) & = & A^{\dagger 2}(t)A^{2}(t)=A^{\dagger 2}A^{2}-2igt\left(2A^{\dagger 3}AB^{3}C-A^{\dagger 2}B^{3}C-2A^{\dagger}A^{3}B^{\dagger 3}C^{\dagger}-A^{2}B^{\dagger 3}C^{\dagger}\right) \\ & + & g^{2}t^{2}\left[24A^{\dagger 2}A^{2}B^{\dagger 3}B^{3}C^{\dagger}C+32A^{\dagger}AB^{\dagger 3}B^{3}C^{\dagger}C+4B^{\dagger 3}B^{3}C^{\dagger}C\right. \\ & - & 36A^{\dagger 3}A^{3}B^{\dagger 2}B^{2}C^{\dagger}C-72A^{\dagger 3}A^{3}B^{\dagger}BC^{\dagger}C-18A^{\dagger 2}A^{2}B^{\dagger 2}B^{2}C^{\dagger}C \\ & - & 36A^{\dagger 2}A^{2}B^{\dagger}BC^{\dagger}C-4A^{\dagger 3}A^{3}B^{\dagger 3}B^{3}-36A^{\dagger 3}A^{3}B^{\dagger 2}B^{2} \\ & - & 72A^{\dagger 3}A^{3}B^{\dagger}B-2A^{\dagger 2}A^{2}B^{\dagger 3}B^{3}-18A^{\dagger 2}A^{2}B^{\dagger 2}B^{2} \\ & - & 4A^{\dagger 4}B^{6}C^{2}-4A^{4}B^{\dagger 6}C^{\dagger 2}-36A^{\dagger 2}A^{2}B^{\dagger}B \\ & - & \left.24A^{\dagger 3}A^{3}C^{\dagger}C-12A^{\dagger 2}A^{2}C^{\dagger}C-24A^{\dagger 3}A^{3}-12A^{\dagger 2}A^{2}\right]. \end{array} \tag{18}$$

In the present study, we have taken all the expectations with respect to $|\alpha>|0>|0>$ for simplification. This assumption physically means that initially a coherent state (say, a laser) is used as pump and before the interaction of the pump with atom, there was no photon in signal mode (b) or stokes mode (c). Thus the pump interacts with the atom and causes excitation followed by emissions. Now from (17) and (18), we have

$$\langle N(t)\rangle^{2}=|\alpha|^{4}-24g^{2}t^{2}|\alpha|^{6}. \tag{19}$$

$$\left\langle N^{2}(t)\right\rangle=|\alpha|^{4}-g^{2}t^{2}\left(24|\alpha|^{6}+12|\alpha|^{4}\right) \tag{20}$$

where $A|\alpha>=\alpha|\alpha>$. Now by using (19) and (20) we can show that the six wave mixing process satisfies the criterion of antibunching (5) since:

$$\begin{array}{lcl} d(1) & = & \left\langle N^{(2)}(t)\right\rangle-\langle N(t)\rangle^{2} \\ & = & \left[|\alpha|^{4}+g^{2}t^{2}(-24|\alpha|^{6}-12|\alpha|^{4})\right]-\left[|\alpha|^{4}-24g^{2}t^{2}|\alpha|^{6}\right] \\ & = & -12g^{2}t^{2}|\alpha|^{4}. \end{array} \tag{21}$$

From the last equation it is clear that $d(1)$ is always negative, i.e. it always shows usual antibunching. Essentially, this is a nonclassical state but mere satisfaction of nonclassicality or antibunching is not enough because we are looking for HOA. Let us see what happens in the next higher order.

For the study of second order of antibunching, $A^{3}(t)$ can be obtained by using [15] and operator ordering techniques:

$$\begin{array}{lcl} A^{3}(t) & = & A^{3}-2igt\left(3A^{\dagger}A^{2}B^{3}C+3AB^{3}C\right) \\ & + & g^{2}t^{2}\left[6A^{3}B^{\dagger 3}B^{3}C^{\dagger}C-27A^{\dagger}A^{4}B^{\dagger 2}B^{2}C^{\dagger}C-54A^{\dagger}A^{4}B^{\dagger}BC^{\dagger}C\right. \\ & - & 54A^{3}B^{\dagger}BC^{\dagger}C-27A^{3}B^{\dagger 2}B^{2}C^{\dagger}C-3A^{\dagger}A^{4}B^{\dagger 3}B^{3}-27A^{\dagger}A^{4}B^{\dagger 2}B^{2}-54A^{\dagger}A^{4}B^{\dagger}B \\ & - & 3A^{3}B^{\dagger 3}B^{3}-27A^{3}B^{\dagger 2}B^{2}-18A^{\dagger}A^{4}C^{\dagger}C-18A^{3}C^{\dagger}C \\ & - & \left.54A^{3}B^{\dagger}B-12A^{\dagger 2}AB^{6}C^{2}-12A^{\dagger}B^{6}C^{2}-18A^{\dagger}A^{4}-18A^{3}\right] \end{array} \tag{22}$$

Then $A^{\dagger 3}(t)$ can simply be written as,

$$\begin{array}{lcl} A^{\dagger 3}(t) & = & A^{\dagger 3}+2igt\left(3A^{\dagger 2}AB^{\dagger 3}C^{\dagger}+3A^{\dagger}B^{\dagger 3}C^{\dagger}\right) \\ & + & g^{2}t^{2}\left[6A^{\dagger 3}B^{\dagger 3}B^{3}C^{\dagger}C-27A^{\dagger 4}AB^{\dagger 2}B^{2}C^{\dagger}C-54A^{\dagger 4}AB^{\dagger}BC^{\dagger}C\right. \\ & - & 54A^{\dagger 3}B^{\dagger}BC^{\dagger}C-27A^{\dagger 3}B^{\dagger 2}B^{2}C^{\dagger}C-3A^{\dagger 4}AB^{\dagger 3}B^{3}-27A^{\dagger 4}AB^{\dagger 2}B^{2}-54A^{\dagger 4}AB^{\dagger}B \\ & - & 3A^{\dagger 3}B^{\dagger 3}B^{3}-27A^{\dagger 3}B^{\dagger 2}B^{2}-18A^{\dagger 4}AC^{\dagger}C-18A^{\dagger 3}C^{\dagger}C \\ & - & \left.54A^{\dagger 3}B^{\dagger}B-12A^{\dagger}A^{2}B^{\dagger 6}C^{\dagger 2}-12AB^{\dagger 6}C^{\dagger 2}-18A^{\dagger 4}A-18A^{\dagger 3}\right] \end{array} \tag{23}$$

Last two equations can be used to calculate the third factorial moment ($N^{(3)}(t)$) of the number operator N as

$$
\begin{aligned}
N^{(3)}(t) &= A^{\dagger 3}A^3 - 2igt\left(3A^{\dagger 4}A^2B^3C + 3A^{\dagger 3}AB^3C - 3A^{\dagger 2}A^4B^{\dagger 3}C^{\dagger} - 3A^{\dagger}A^3B^{\dagger 3}C^{\dagger}\right) \\
&+ g^2t^2\left[48A^{\dagger 3}A^3B^{\dagger 3}B^3C^{\dagger}C - 54A^{\dagger 4}A^4B^{\dagger 2}B^2C^{\dagger}C - 54A^{\dagger 3}A^3B^{\dagger 2}B^2C^{\dagger}C\right. \\
&- 108A^{\dagger 3}A^3B^{\dagger}BC^{\dagger}C + 108A^{\dagger 2}A^2B^{\dagger 3}B^3C^{\dagger}C + 36A^{\dagger}AB^{\dagger 3}B^3C^{\dagger}C \\
&- 108A^{\dagger 4}A^4B^{\dagger}BC^{\dagger}C - 54A^{\dagger 4}A^4B^{\dagger 2}B^2 - 108A^{\dagger 4}A^4B^{\dagger}B - 6A^{\dagger 3}A^3B^{\dagger 3}B^3 \\
&- 6A^{\dagger 4}A^4B^{\dagger 3}B^3 - 54A^{\dagger 3}A^3B^{\dagger 2}B^2 - 108A^{\dagger 3}A^3B^{\dagger}B - 12A^{\dagger 5}AB^6C^2 - 12A^{\dagger}A^5B^{\dagger 6}C^{\dagger 2} \\
&- \left. 12A^{\dagger 4}B^6C^2 - 12A^4B^{\dagger 6}C^{\dagger 2} - 36A^{\dagger 3}A^3C^{\dagger}C - 36A^{\dagger 4}A^4C^{\dagger}C - 36A^{\dagger 4}A^4 - 36A^{\dagger 3}A^3\right].
\end{aligned} \tag{24}
$$

Taking the expectation value with respect to the intial state we obtain

$$
\left\langle N^{(3)}(t)\right\rangle = |\alpha|^6 - g^2t^2\left(36|\alpha|^8 + 36|\alpha|^6\right). \tag{25}
$$

On the other hand,

$$
\langle N(t)\rangle^3 = |\alpha|^6 - 36g^2t^2|\alpha|^8 \tag{26}
$$

By using the last two equations we can see that the pump mode of six wave mixing process satisfy the criteria of antibunching of second order (5). Since,

$$
\begin{aligned}
d(2) &= \left\langle N^{(3)}(t)\right\rangle - \langle N(t)\rangle^3 \\
&= \left[|\alpha|^6 - g^2t^2(36|\alpha|^8 + 36|\alpha|^6)\right] - \left[|\alpha|^6 - 36g^2t^2|\alpha|^8\right] \\
&= -36g^2t^2|\alpha|^6
\end{aligned} \tag{27}
$$

is always negative.

3 Four wave mixing process

Similarly, four wave mixing may also happen in different ways. One way is that in which two photon of frequency ω_1 are absorbed (as pump photon) and one photon of frequency ω_2 and another of frequency ω_3 are emitted. The Hamiltonian representing this particular four wave mixing process is

$$
H = a^{\dagger}a\omega_1 + b^{\dagger}b\omega_2 + c^{\dagger}c\omega_3 + g(a^{\dagger 2}bc + a^2b^{\dagger}c^{\dagger}). \tag{28}
$$

Following the same prescription as it is used in six wave case we can write the solution as

$$
A(t) = A - 2igtA^{\dagger}BC + \frac{g^2t^2}{2!}[4AB^{\dagger}BC^{\dagger}C - 2A^{\dagger}A^2B^{\dagger}B - 2A^{\dagger}A^2C^{\dagger}C - 2A^{\dagger}A^2] \tag{29}
$$

or

$$
A(t) = A - 2igtA^{\dagger}BC + g^2t^2[2AN_BN_c - N_AAN_B - N_AAN_C - N_AA]. \tag{30}
$$

The respective values of the first order antibunching ($d(1)$) and second order antibunching ($d(2)$) can similarly be calculated as we have done for six wave mixing and that yields

$$
\begin{aligned}
d(1) &= \left\langle N^{(2)}(t)\right\rangle - \langle N(t)\rangle^2 \\
&= \left[|\alpha|^4 + g^2t^2(-4|\alpha|^6 - 2|\alpha|^4)\right] - \left[|\alpha|^4 - 4g^2t^2|\alpha|^6\right] \\
&= -2g^2t^2|\alpha|^4.
\end{aligned} \tag{31}
$$

and

$$
\begin{aligned}
d(2) &= \left\langle N^{(3)}(t)\right\rangle - \langle N(t)\rangle^3 \\
&= \left[|\alpha|^6 - g^2t^2(6|\alpha|^8 + 6|\alpha|^6)\right] - \left[|\alpha|^6 - 6g^2t^2|\alpha|^8\right] \\
&= -6g^2t^2|\alpha|^6
\end{aligned} \tag{32}
$$

which are negative and thus they satisfy our criterion for antibunching and HOA respectively.

4 Second harmonic generation

The similar procedure can be repeated for the second harmonic generation process whose Hamiltonian is

$$H = \hbar\omega N_1 + 2\hbar\omega N_2 + hg\left(a_2^\dagger a_1^2 + a_1^{\dagger 2} a_2\right). \tag{33}$$

The second order expression of the time evolution of annihilation operator in pump mode of second harmonic generation is

$$A(t) = a_1 - 2igta_1^\dagger a_2 + 2g^2t^2\left(a_2^\dagger a_2 a_1 - \frac{1}{2}a_1^\dagger a_1^2\right). \tag{34}$$

Using the last equation along with the method used in section 2 we obtain

$$\begin{aligned} d(1) &= \left\langle N^{(2)}(t)\right\rangle - \langle N(t)\rangle^2 \\ &= \left[|\alpha|^4 + g^2t^2(-4|\alpha|^6 - 2|\alpha|^4)\right] - \left[|\alpha|^4 - 4g^2t^2|\alpha|^6\right] \\ &= -2g^2t^2|\alpha|^4 \end{aligned} \tag{35}$$

and

$$\begin{aligned} d(2) &= \left\langle N^{(3)}(t)\right\rangle - \langle N(t)\rangle^3 \\ &= \left[|\alpha|^6 - g^2t^2(6|\alpha|^8 + 6|\alpha|^6)\right] - \left[|\alpha|^6 - 6g^2t^2|\alpha|^8\right] \\ &= -6g^2t^2|\alpha|^6 \end{aligned} \tag{36}$$

which are both negative and hence satisfies the criterion (5) for antibunching and HOA respectively.

5 Conclusions:

From (27, 32 and 36), it is clear that all the physical systems selected for the present study show second order antibunching i.e. higher order sub-poissonian photon statistics. Thus the present work strongly establishes the fact that HOA is not a rare phenomenon. The physical systems studied in the present paper are simple and easily achievable in laboratories and thus it opens up the possibility of experimental observation of HOA. In case of interaction of intense electromagnetic field with third order nonlinear medium it was reported [11] that the degree of antibunching ($d(l)$) can be tuned because they depend strongly on the phase of the input field which can be tuned. This is not the case with any of the physical systems studied in the present work. It is also clear that the higher order antibunching would not have been observed if we would have considered first order operator solutions (first order in g), on the other hand, if we use second order operator solutions then the depth of nonclassicality is found to increase monotonically with the increase of input photon number ($|\alpha|^2$). Possibly this monotonic increment will be ceased by the higher order perturbation terms. It is also observed that if we assume that the anharmonic constant and number of photon initially present in the pump mode are same for all three cases then the depth of nonclassicality is same in four wave mixing and second harmonic generation process and its more in six wave mixing process.

The prescription followed in the present work is easy and straight forward and it can be used to study the possibilities of observing higher order antibunching in other physical systems. Thus it opens up the possibility of studying higher order nonclassical effects from a new perspective. This is also important from the application point of view because any probabilistic single photon source used for quantum cryptography has to satisfy the condition for higher order antibunching. Therefore, the simple prescription followed in the present work may help us to compare the existing sources of single photon.

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